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Scaling relation for the superfluid density in cuprates - origins and limits

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A universal scaling relation, $\rho_s \propto \sigma(T_c) \times T_c$ has been reported by Homes *et al.* (Nature (London) **430**, 539 (2004)) where ρ_s is the superfluid density and $\sigma(T)$ is the DC conductivity. The relation was shown to apply to both *c*-axis and in-plane dynamics for high-T_c superconductors as well as to the more conventional superconductors Nb and Pb, suggesting common physics in these systems. We show quantitatively that the scaling behavior has several possible origins including, marginal Fermi-liquid behavior, Josephson coupling, dirty-limit superconductivity and unitary impurity scattering for a *d*-wave order parameter. However, the relation breaks down seriously in overdoped cuprates, and possibly even at lower doping.

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The absence of a recognized theory of high-T_c superconductors (HTS) has led to a search for universal relationships that might provide a guide to the essential physics for HTS. The Uemura relation[1, 2] is one such scaling relation, namely $\rho_s(0) \propto T_c$, where T_c is the superconducting (SC) transition temperature and $\rho_s(0) = \lambda_{ab}^{-2} = \mu_0 e^2 n_s/m^*$ is the superfluid density. Here n_s is the density of SC electrons, m^* is their effective mass and λ_{ab} is the in-plane London penetration depth. The Uemura relation has been invoked in support of Bose-Einstein condensation of real-space pairs[2, 3] and is generally discussed as a test of theoretical models[4]. However, there is increasing recognition that this relation is an oversimplification[5, 6], and it breaks down on the overdoped side of the SC phase curve[7].

Recently, a new scaling relation, $\rho_s \propto \sigma(T_c) \times T_c$ was reported by Homes *et al.*[8] where $\sigma(T)$ is the DC conductivity. This was shown to apply over five orders of magnitude including both *c*-axis and in-plane dynamics for HTS as well as to the conventional superconductors Nb and Pb. The authors suggested this relation may provide new insights into the origins of SC in HTS materials. We examine this relation and show that it is a natural consequence of several quite different but well-understood mechanisms, including dirty-limit conventional SC (Pb and Nb), Josephson coupling along the *c*-axis, marginal Fermi-liquid behavior (optimal and underdoped HTS), in-plane Josephson-coupled granular SC (strongly underdoped HTS) and Abrikosov-Gor'kov *d*-wave pair-breaking for non-magnetic scatterers. Since we submitted this work Homes *et al.*[9] have reported some overlapping results but here we examine the doping and impurity dependence of this relation showing that, in overdoped HTS, it breaks down seriously. It is perhaps significant that, here, the cuprates progress towards more conventional SC and normal state (NS) behavior.

Because $\lambda^{-2} = \mu_0 e^2 n_s/m^*$ and $\sigma = ne^2\tau/m^*$ the correlation is equivalent to a relationship between scattering rate $1/\tau$ and T_c given by $\hbar/\tau = 2.7 \pm 0.5 \times k_B T_c$. Here we assume that all the spectral weight associated with the free carriers condenses into the δ-function at ω=0, i.e. n_s=n. In fact, for underdoped cuprates some spectral weight remains at finite frequency[10]. However, we assume that this weight is related to some other excitation, different from the free carriers, which does not contribute to $\sigma(\omega$=0), ρ_s or the low-T specific heat.

In considering possible origins for this relation between $\hbar/\tau$ and $k_B T_c$ we note that it is a direct prediction from Marginal-Fermi-liquid theory, where[11, 12]

$$\hbar/\tau \approx \pi k_B T + \hbar\omega. \qquad (1)$$

At low frequency, the scaling relation is almost exactly recovered provided τ is evaluated at T_c, as required.

That Nb and Pb fit the scaling line is surprising, but it is readily shown that for classical SC in the dirty limit ρ_s is, again, proportional to $\sigma(T_c) \times T_c$. For a mean-free path, ℓ, and a BCS coherence length, $\xi_0 = \hbar v_F/(\pi\Delta)$, the effective penetration depth is given by[13]

$$\lambda_{eff} = \lambda_L(1 + \xi_0/\ell)^{1/2} \approx \lambda_L(\xi_0/\ell)^{1/2}. \qquad (2)$$

Taking $\ell = v_F\tau$ and $\Delta = 1.76 k_B T_c$, we find that ρ_s is a factor of two higher than the scaling line ($\hbar/\tau = 5.4 \times k_B T_c$). In the clean limit there will be large deviations below the scaling line because ρ_s remains constant while $\sigma(T_c) \times T_c$ can be extremely large. Using data for Pb alloys[14] and for Nb alloys[15] we find that, for the Pb sample on the Homes scaling line, $\xi_0/\ell = 1.2$ while the two Nb samples have $\xi_0/\ell = 0.6$ and 2.1. From eq. (2) it can be seen that in the intermediate situation where $\xi_0/\ell \approx 1$ the above factor of two is compensated. The apparent scaling here is understandable but fortuitous.

Homes *et al*[8] consider Josephson coupling along the *c*-axis and show the proportionality $\rho_s \propto \sigma \times T_c$ but do not

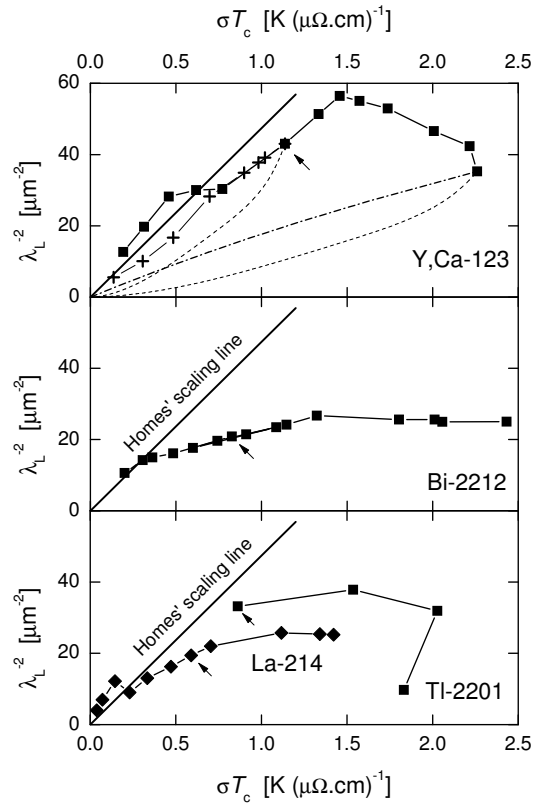

FIG. 1: Superfluid density, ρ_s versus $\sigma(T_c) \times T_c$ for (a) $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$, (b) $Bi_2Sr_2CaCu_2O_{8+\delta}$ and (c) $Tl_2Ba_2CuO_{6+\delta}$ (overdoped only) and $La_{2-x}Sr_xCuO_4$. Arrows indicate optimal doping. In (a) the effects of Zn-induced pairbreaking are shown (crosses) along with the calculated A-G pairbreaking effects for constant σ (dashed curve) and for σ reduced by impurity scattering (dash-dot curve). The Homes' data lie within $\sim \pm 25\%$ of the scaling line.

evaluate the proportionality constant. The c-axis penetration depth, λ_c is expressed in terms of the Josephson current density, J_c as $\lambda_c^2 = \hbar/(2\mu_0 deJ_c)$ where d is the spacing between SC planes and, for low temperatures, $J_c = \pi\Delta/(2eR_n)$[16]. $R_n = d/\sigma_c$ is the NS tunnelling resistance between layers. For a d-wave gap we may assume that the effective gap parameter, $\Delta_{eff} = \Delta_0/\sqrt{2}$ where $\triangle_0$ ($\approx 2.4k_BT_c$) is the maximum d-wave gap near $(\pi, 0)$. Thus $\lambda_c^{-2} = 6.79(\pi^2/\hbar c^2)k_BT_c\sigma_c$ giving the relation $\hbar/\tau = 2.23\times k_BT_c$. If instead we consider a simple rectangular I-V Josephson characteristic, then $I_c = 2\Delta_0/eR_n$ and the relation becomes $\hbar/\tau = 2.83 \times k_BT_c$, again in excellent agreement with the scaling curve.

Thus, whether we consider Josephson coupling, dirty-limit SC or marginal-Fermi-liquid SC we recover the observed scaling behavior. Now we turn to the experimental data for the doping dependence of the in-plane dynamics.

In Fig. 1 the filled squares and diamonds show values of $\rho_s(0)$ plotted versus $\sigma(T_c) \times T_c$ for four different HTS materials. The samples, technique for measuring $\rho_s(0)$ and sample-type are as follows: (a) $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ (transverse-field muon spin relaxation - μSR, polycrystalline), (b) $Bi_2Sr_2CaCu_2O_{8+\delta}$ (field-dependent specific heat, polycrystalline), and (c) $Tl_2Ba_2CuO_{6+\delta}$ (μSR, polycrystalline) with $La_{2-x}Sr_xCuO_4$ (ac-susceptibility, oriented powders). The $\rho_s(0)$ data for $Tl_2Ba_2CuO_{6+\delta}$ are from ref. [7] and the remainder are from references in [5]. The techniques noted are well established except for the specific-heat method. But $\rho_s(0)$ has also recently been measured for $Bi_2Sr_2CaCu_2O_{8+\delta}$ using ac-susceptibility[17]. Apart from the heavily underdoped region, the data are in excellent agreement with those used here from specific-heat.

The in-plane $\sigma(T)$ data in Fig. 1 are from DC transport measurements and for these it is essential to use single-crystal measurements. These are available for all cited samples except $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ for which we have used thin-film data from Fisher[18], supported by more recent data from Naqib *et al.*[19]. Sources for the remaining samples are: $Bi_2Sr_2CaCu_2O_{8+\delta}$ from Watanabe *et al.*[20]; for $Tl_2Ba_2CuO_{6+\delta}$ from Tyler and coworkers[21] and for $La_{2-x}Sr_xCuO_4$ from Ando and coworkers[22]. In all cases we evaluated $\sigma(T_c)$ at the reported doping states, fitted to an appropriate function of doping and interpolated to determine $\sigma(T_c)$ at the doping state relevant to the available ρ_s data. For $Bi_2Sr_2CaCu_2O_{8+\delta}$, the $\sigma(T_c)$ data had to be extrapolated to higher doping levels. In Fig. 1 optimal doping (maximal T_c) is indicated by the arrows and critical doping, where the pseudogap closes, is the point of maximum ρ_s[5]. The overdoped region lies further to the right.

Our main conclusion is that each compound exhibits a large departure from the "universal" scaling line in the overdoped region. In the optimal and underdoped regions, where we can directly compare with Homes *et al*, the situation is less clear. In some cases our data differs markedly from theirs which severely overestimates ρ_s for $Bi_2Sr_2CaCu_2O_8$ relative to data from specific heat, ac susceptibility and μSR and underestimates ρ_s for $Tl_2Ba_2CuO_6$. For this compound, and for $La_{2-x}Sr_xCuO_4$, their $\sigma(T_c)$ value is about half that observed from DC transport in single crystals[21]. The source of these discrepancies may possibly be attributable to the extrapolation to $\omega = 0$ and $\omega = \infty$ required for the Kramers-Kronig transformation of the optical data.

Bearing in mind these differences our underdoped results show a consistent trend. For $Bi_2Sr_2CaCu_2O_8$, $Tl_2Ba_2CuO_6$ and $La_{2-x}Sr_xCuO_4$ the scaling line is not reached until near, or below, $1/8^{th}$ doping. On the other hand $Y_{1-x}Ca_xBa_2Cu_3O_7$, though not linear, roughly follows the trend of the scaling line across the entire underdoped region. However, there are two reasons why this could misrepresent the situation. Firstly, compared with other bilayer cuprates with similar T_c this compound generally exhibits a higher superfluid density arising from

the contribution of the chain layer. This may be approximately balanced in the scaling plot by the additional conductivity also arising from the chains and so we put this effect to one side. But secondly, the conductivity here is for thin films and thus is probably diminished relative to single-crystal data. All the data points may lie further to the right, perhaps by a factor of the order of 1.5. Then the behavior would be rather similar to that shown by $Bi_2Sr_2CaCu_2O_{8+\delta}$ and $La_{2-x}Sr_xCuO_4$. It may be that there is no correlation with the scaling line until the lowest doping state around $p = 1/8$ and lower.

What can we conclude from this? One implication may be that in this heavily underdoped region the in-plane physics is rather similar to the out-of-plane physics, namely that of Josephson-coupled SC domains. Here the electronic state is probably granular, with SC patches separated by insulating Josephson barriers. Such a model has been proposed even for optimally doped cuprates and STM suggests the presence of nanoscale inhomogeneity[23, 24]. However this has been questioned[25], and in particular the electronic state seen from the perspective of NMR and specific heat seems to be homogeneous for $p > 0.12$[26]. Below that doping state these techniques reveal the presence of normal quasiparticles[27]. Here the electronic state definitely appears to become spatially heterogeneous.

We turn now to ask the question as to whether the universal scaling relation may also apply in the case of impurity scattering. This behavior can be understood in terms of generally-accepted theories for the pair-breaking effects of Zn impurities, namely the suppression of T_c by non-magnetic scattering, which for a d-wave superconductor suppresses T_c according to the Abrikosov-Gor'kov (A-G) formula and ρ_s according to the work of Maki and coworkers[28]. When we add in the effects of scattering on $\sigma(T)$ (described by Matthiessen's rule) this roughly reproduces the observed scaling behavior for optimally-doped samples but, again, we expect large deviations for overdoped samples for which conductivity data as a function of Zn doping are not yet available. The theory can be developed along the following lines.

For a non-magnetic scatterer d-wave SC is rapidly suppressed and the effects on T_c, ρ_s and σ as a function of impurity concentration, y, are governed by the density of states (DOS) at the Fermi level. These effects have been shown to be fully consistent with thermodynamic data[29]. The reduction in T_c is given by:

$$-\ln(T_c/T_{c0}) = \psi[1/2 + \Gamma/\Gamma_c] - \psi[1/2], \qquad (3)$$

where $\psi[x]$ is the digamma function, $T_{c0} = T_c(y{=}0)$. For unitary scattering, $\Gamma = n_i/\pi g(E_F)$ is the pair-breaking scattering rate and Γ_c is its critical value for which T_c is suppressed to zero. Here $g(E_F)$ is the DOS per spin, $n_i(= \alpha y/abc)$ is the density of impurities, α is the number of CuO_2 planes per unit cell and a, b and c are the lattice parameters. As before[29], we adopt the strong-coupling scenario $\Gamma_c = 1.65 k_B T_{c0}$ and determine $g(E_F)$ from the electronic specific heat coefficient, γ, using $\gamma = (2/3)\pi^2 k_B^2 g(E_F)$. The following linearized form of the A-G equation is valid up to about $(2/3)\Gamma_c$:

$$T_c/T_{c0} = 1 - 0.69\Gamma/\Gamma_c = 1 - (0.86\alpha R/\gamma T_{c0}) \times y, \qquad (4)$$

where $R = N_A k_B$. In the overdoped region γ is constant, independent of temperature and doping, so that the slope, $\partial T_c/\partial y$, in eqns. (3) or (4) remains constant. As a practical measure[29], the value of γ was taken as S/T evaluated at T_c, where S is the electronic entropy. This is just the average of $\gamma(T)$ between $T = 0$ and T_c.

The calculation of the reduction in superfluid density is more complex and is carried out numerically[28]. However, we find an excellent approximation to within a few percent across the entire pair-breaking range using:

$$\rho_s/\rho_{s0} \approx \frac{T_c/T_{c0}}{1 + 1.7(1 - T_c/T_{c0})}. \qquad (5)$$

This non-linear relation reflects the fact that ρ_s is initially suppressed much faster than T_c[5]. Eqns. (3) and (5) are universal relations and if impurity scattering were to have no effect on σ then the effect of Zn substitution would be given by the dashed A-G curves shown in Fig. 1(a) for the optimal and most overdoped $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$.

In fact, the resistivity is increased by impurity scattering according to a linear Matthiessen law. The resultant decrease in $\sigma(T)$ seems to effectively linearize the simple A-G curve in the scaling plot. We illustrate this using the experimental data shown by the crosses in Fig. 1. These are data for an optimally-doped sample where the reduction in ρ_s and T_c is from μSR measurements by Bernhard *et al.*[31], and the $\sigma(T)$ data for Zn substitution at the same doping state are from thin-film samples studied by Naqib *et al*[19]. These trend back linearly to the origin in reasonably good agreement with the scaling curve. We now seek to justify this quantitatively.

The residual planar resistivity arising from impurity scattering is given by[30] $4(\hbar/e^2)(n_i/n)sin^2\delta_0$ where δ_0 is the s-wave scattering phase shift which, in the unitary limit, we take to be $\pi/2$. Thus the total resistivity is $m^*/(ne^2\tau) + 4c(\hbar/\alpha e^2)(n_i/n)$ and σ becomes

$$\sigma = \frac{ne^2\tau/m^*}{1 + (4\hbar\tau/abm^*) \times y} \qquad (6)$$

where τ is evaluated at $T_c(y)$.

There are two impurity effects to be considered. Firstly, there is the Matthiessen term in the denominator of eqn.(6) and, secondly, $\sigma(T)$ must be evaluated *at* T_c which is itself reduced by scattering. This second effect is accommodated by the fact that, in eqn. (7) below, the only two variables in the coefficients are τ and T_c and they appear as the product τT_c. Thus, provided the

material lies on the scaling curve in the absence of impurities, this product is a constant. Proceeding, the explicit variable y in eqn. (6) may be eliminated by substituting from eqn. (4) to give:

$$\sigma(y)T_c(y) = \frac{\sigma(0)T_{c0} \times (T_c(y)/T_{c0})}{1 + (\beta - 1)(1 - T_c(y)/T_{c0})}. \qquad (7)$$

where $\beta = 4\hbar\gamma\tau_0 T_{c0}/0.86\alpha Rabm^*$ and $\tau_0 = \tau$ evaluated at $T_c(y = 0)$. Taking $m^* \approx 2m_e$, as observed for nodal quasiparticles[33], we have only to note that the right side of eqn.(7) is of the same form as eqn.(5) with the coefficient $(\beta - 1) = 1.9$. Thus

$$\sigma(y)T_c(y)/\rho_s(y) \approx \sigma(0)T_{c0}/\rho_{s0}, \qquad (8)$$

and the impurity scattering data remain on the scaling curve if, in the first instance, it lies on the curve in the absence of impurity scattering. This seems to explain the crosses shown in Fig. 1. This calculation shows that, with underdoping, the curve would steepen due to the further opening of the pseudogap (and the associated reduced entropy at T_c) while with overdoping it would flatten (due to the closing of the pseudogap). It would be nice to test this but we do not have combined $\rho_s(y)$ and $\sigma(y)$ data as a function of Zn concentration for heavily overdoped samples. Nevertheless, we have calculated the expected effect of Zn substitution using eqn. (7) and this is plotted in Fig. 1 by the dotted line. This shows a fairly linear behavior but which is displaced far from the "universal" scaling curve.

In conclusion, we have examined the scaling relation, $\rho_s \propto \sigma(T_c) \times T_c$, and find this is equivalent to the relation $\hbar/\tau = 2.7 \pm 0.5 \times k_B T_c$ which could equally arise from (i) conventional dirty-limit SC, (ii) marginal-Fermi-liquid behaviour, (iii) Josephson-coupling along the c-axis of the CuO_2 planes, or (iv) unitary-limit impurity scattering. We conclude that the Pb and Nb samples correlate because these samples are near the dirty limit. Interestingly, the Homes scaling line corresponds to the mean free path just above T_c being approximately $2 \times \xi_0$ (using the maximum gap $\approx 2.38 k_B T_c$ for a weak-coupling d-wave SC). Even more intriguingly, in terms of standard microscopic BCS theory[32], it corresponds to the Gor'kov kernel having a range ξ_G near T_c equal to the mean free path just above T_c.

When we examine the doping dependence of the in-plane ρ_s and $\sigma(T_c) \times T_c$ we find a total breakdown of the scaling relation across the overdoped region. This shows that the mean free path at T_c is now much larger than both of the above-noted coherence lengths. This breakdown may extend into the underdoped region, allowing for the fact that the only exception is for *thin-film* $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$. The recovery of the scaling behavior at very low doping may arise from similar physics to c-axis Josephson coupling, namely, weak-linked patches of superconducting domains in the inhomogeneous strongly-underdoped state.

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